# ´Waste-to-Energy' Fuel Cell Systems

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In this paper, a review of the different possible gas and solid fuels for solid oxide fuel cells (SOFCs) is presented. Much research has been performed with gaseous fuels in SOFCs. On the contrary, much work remains on the direct use of solid fuels in SOFCs to overcome all the technical challenges that these systems present. The challenges are even greater when the use of complex solid waste is considered. However, the development of efficient and sustainable energy systems that can operate with waste is of general interest to the energy sector and the environment, as waste management is of major concern. In particular, the re-utilisation or disposal of plastics is of great importance due to their worldwide usage and their slow degradation. The use of an untreated wood and polystyrene mixture in an electrolyte-supported fuel cell with a NiO-YSZ anode and LSM-based cathode was also investigated in this work.

## Introduction

The achievement of a sustainable and renewable energy supply is one of the main challenges that our society faces. An equally important problem is the management of waste. Landfill sites, incineration or disposal in the ocean are not sustainable long-term solutions, especially from an environmental perspective. An ideal scenario would be to solve both issues simultaneously, by developing more efficient and environmentally-friendly 'waste-to-energy' systems. Fuel cells are a clean method of energy generation that can directly convert the chemical energy of a fuel into electricity and heat with high efficiency. Their efficiency is generally higher than from combustion of existing fossil fuels and can be further improved using Combined Heat and Power (CHP) systems (1). In a CHP, the gas exhaust from the fuel cell, which can contain unconverted $CH_4$ or $H_2$, can be used in an after-burner, gas turbine or steam turbine to generate heat or mechanical power, increasing the overall system efficiency. The overall thermal and electrical efficiency of an integrated SOFC system (with gas or steam turbine) can reach up to 90% (2).

SOFCs present the advantage of being able to operate at high temperature (500 – 950 °C), which enables the use of a variety of fuel sources, *i.e.* hydrogen-, carbon- (such as CO or hydrocarbons) and nitrogen-based (such as ammonia) feedstocks (3). Hydrogen is regarded as the ideal fuel. However, the difficulties associated with its production, compression, storage and distribution, means that alternative carbon or nitrogen-based fuels should be considered.

More specifically, SOFCs can use gaseous and solid carbon feedstocks. Both feedstocks can be produced from fossil fuels or from renewable resources, such as biomass or waste sources. The limited availability of fossil fuels, their increasing prices, environmental impacts and issues related to the security of energy supply are driving forces towards the utilisation of more sustainable resources (4). Therefore, there is increased interest in potential fuels from renewable resources for SOFCs (5-7). However, the use of such fuels implies new challenges for this technology, mainly due to their variety and variation of compositions and the presence of impurities (7). Biomass-derived fuels are an interesting option as they provide a neutral $CO_2$ circle. The term biomass refers to natural organic materials associated with living organisms, including terrestrial and marine vegetable matter, animal tissue and manure. Therefore, its definition can extend to include waste streams such as municipal solid wastes (MSW), animal waste, food waste or aquatic plants such as algae. Ideally, unutilised waste streams should be prioritised over resources which already have a market, such as crops used in food production, as this can reduce the amount of waste disposed (5).

The most common gaseous fuels derived from fossil feedstocks are: (i) natural gas (NG, a hydrocarbon gas mixture, mainly comprising of $CH_4$ but can also contain other higher alkanes, $CO_2$, $N_2$, $H_2S$ or He), (ii) $H_2$ produced from steam methane reforming of natural gas and (iii) other gas mixtures produced by external gasification of complex higher hydrocarbons such as gasoline, alcohols, diesel, kerosene and other liquid-based fuels (8). Some of the main gaseous feedstocks derived from biomass are: (i) biogas ($CH_4$/$CO_2$), biohydrogen ($H_2$/$CO_2$) and (iii) gas mixtures from gasification processes and industrial waste gases (3). A detailed review of the possible pathways to produce fuel gases for SOFCs from biomass/waste has been carried out by Archer and Steinberger-Wilckens. According to their work, the biomass/waste conversion techniques can be classified in three main categories: (i) biological (fermentation, anaerobic digestion and metabolic processing), (ii) thermochemical (gasification and supercritical water gasification for gas production or pyrolysis followed by liquefaction for liquid fuels) and (iii) extraction of carbohydrates, lipids and hydrocarbons for alcohol and biodiesel production. Agricultural crop wastes, waste fats and vegetables oils (sources high in carbohydrates and proteins) can be utilised in biological routes to obtain biogas, methane and hydrogen; whereas thermochemical conversion routes use mainly lignocellulosic biomass to produce syngas. Moreover, anaerobic digestion and metabolic routes can use waste residues, such as slurries and wastewater (5). Saadabadi *et al*. have reviewed the biogas production from different types of wastewater streams by anaerobic digestion, the possible cleaning and reforming biogas techniques and the advantages and challenges of integrated anaerobic digestion-SOFC systems (9). Additional processing, such as reforming and/or purification, are normally needed for most of the previously described routes to obtained $H_2$-rich streams that can be used in low temperature SOFCs, except for the metabolic and fermentation routes which directly produce $H_2$ (5).

Independent of the gas fuel source (fossil fuel or biomass-derived), the reformation step, when needed, can be carried out either inside the SOFC, or externally using a reformer unit. Managing the reformation process is quite challenging due to the impacts that it can have on SOFC performance. Steam reforming (SR), dry reforming, non-catalytic partial oxidation (POX) and catalytic partial oxidation (CPOX) are some examples of these possible reforming strategies. Steam reforming involves the catalytic

reaction of hydrocarbon fuels with steam to produce $H_2$, whereas partial oxidation involves the partial combustion of the fuel (8). Steam reforming has been proposed for $CH_4$ reforming of natural gas and syngas. High temperatures (endothermic process) and high steam/carbon ratios are favourable, however the optimum ratios depend on operating conditions. Dry reforming is interesting for mixtures which contain $CO_2$ and $CH_4$, such as biogas, as the reforming agent is already present in the gas mixture. Additionally, the $CO_2$ can prevent carbon deposition by gasification. However, dry reforming is a highly endothermic process and the SOFC needs to be operated at 800-1000 °C to obtain high $CH_4$ conversion. Therefore, if it is carried out internally, it can cause a temperature gradient inside the SOFC that can result in cell cracking. For biogas, as the molar fraction of $CH_4/CO_2$ varies between 1.2 and 1.9, additional $CO_2$ might need to be supplied, for example from the recirculation of the anode exhaust flow. When steam and dry $CH_4$ reforming are carried out externally, a heat source is required to run the reformer (9). Partial oxidation, by nature, produces less $H_2$ per mole of fuel than steam reforming. Also, the use of air as a reactant dilutes the $H_2$ product with $N_2$ and can re-oxidise the anode. Therefore, the use of an external unit is normally preferred. As POX is and exothermic process, it can minimise the required pre-heating of the fuel gas (8). Additionally, there are mixed methods such as autothermal reforming (ATR). With ATR, the POX and SR take place in a reactor by feeding the hydrocarbon source with both steam and air (9).

Minor and trace impurities present in the fuel gas have a significant effect on the performance and durability of SOFCs. Carbon deposition from organic species is one of the main risks. From the inorganic impurities, sulfur in the form of hydrogen sulphide ($H_2S$) is one of the principal impurities studied, as it can poison most catalysts and is corrosive. It can be found in sour gas, biogas or syngas produced from reforming of hydrocarbons and/or carbon (*e.g.* coke); it is also a by-product of many industrial operations such as processing of natural gas, coking and hydrodesulphurization of oil and coal. Zhou *et al.* have summarised the effects of $H_2S$ depending on the fuel gas used, from hydrocarbon ($CH_4$) to syngas ($CO+H_2$) to the use of the pure $H_2S$ as fuel. They have concluded that, given the correct anode catalyst, pure $H_2S$ could be used as a fuel, although more work is required in this area (10). Besides, some studies have found that the presence of $H_2$ increases the sulfur tolerance of SOFCs (3). Less attention has been paid to other inorganic impurities present in vapour phases that can be contained in coal- or biomass-derived syngas or digester gas. Cassidy *et al* have reviewed the impact of these other minor inorganic constituents on different SOFC anodes, the advisable levels in the gas stream and the possible synergetic effects between them or with the anode material. In this work, special attention was paid to (i) P and Se which can be present in coal syngas in the form of phosphine ($PH_3$) and $H_2Se$, AsSe and PbSe, respectively; (ii) Si, As and Sb which can be in the form of $Si(OH)_4$ in coal syngas or siloxanes in biomass-derived fuels, arsine ($AsH_3$), $As_4$ or $As_3Pb$ in coal syngas and $SbO_2H_2$ in coal syngas, respectively; (iii) Hg or Cd that can exit in the form of metal vapour in coal gases and (iv) halogens such as Cl in the form of HCl and $CH_3Cl$ in coal syngas (8).

The direct utilization of solid carbon fuels in SOFCs is technically challenging in many aspects, starting by achieving the feeding of the solid fuel into the fuel cell if the system is to be under continuous operation. These fuel cells work on the same principles as $H_2$-fuelled SOFCs, however the limited contact between the solid fuel particles and the anode-electrolyte interface is one of the limiting factors to power generation (11).

Therefore, one of the most common approaches is to transform the solid fuel into a gaseous form prior to utilisation in the fuel cell. For example, Naqvi *et al.* used a steam gasifier, with the ability for refilling carbon fuel, directly connected to an SOFC, achieving a maximum power density of 181 $mW \cdot cm^{-2}$ at 850 °C with a commercial activated carbon (mixed with $K_2CO_3$) (12). However, other strategies to increase the triple phase boundary (TPB) and accelerate carbon conversion have been investigated. Among them, the use of a molten media such as a carbonate mixture, that can act as a secondary electrolyte and as a gasifying agent simultaneously, and which has led to the development of Hybrid Direct Carbon Fuel Cells (HDCFCs). This strategy has proved to be efficient with different types of complex solid fuels such as mineral coals (13). The use of a gasifying agent, such as $CO_2$, or solid catalysts mixed with the fuel to promote carbon gasification is another common strategy (14, 15). Cao *et al.* have recently published a review about the advances in material development for catalysts/anodes and novel designs according to the different types of direct carbon fuel cells (DCFCs). Most of the work carried out with solid fuels is focused on the use of activated carbon, graphite, carbon black, carbon fibres, pyrolyzed medium density fibreboard (pMDF), mineral coals and chars or carbon produced from different biomass/waste sources, such as paper, bamboo, pine, *etc.* (16). However, very few studies can be found on plastics, which are of major concern for the pollution on land and in water. Plastics are one of the major components of MSW, up to 7 wt.% and greater by volume (17). In a recent study, Cai *et al.* investigated the use of gases from the pyrolysis-gasification of polypropylene (PP) in an SOFC with a Ni-Al-Mg reforming catalyst above the cell. The pyrolysis-gasification step of a fixed amount of PP was carried out in a separate reactor and, therefore, the solid and liquid residues were not interfering with the SOFC operation. Above 480 °C the PP fully decomposed into gases, with only 2 wt.% of solid remaining from the inorganic additives incorporated into plastics to improve their mechanical strength and processability. Higher operation temperatures were found more favourable as fewer proportions of C2-C4 gases were present (18).

In this paper, a wood and plastic (polystyrene-PS) waste-fuelled SOFC system was investigated. An electrolyte-supported SOFC with a NiO-YSZ anode and an LSM-based cathode was used for these initial studies. The results demonstrate the feasibility of using this waste mixture without any pre-processing, pre-gasification or the aid of secondary electrolytes. The anode electrode and solid fuel were fully characterised before and after electrochemical testing *via* powder x-ray diffraction, electron microscopy and, in the case of the solid waste, elemental analysis.

## Experimental

### Fuel Cell Fabrication

Electrolyte-supported yttria-stabilised zirconia (8YSZ, Pi-kem) button cells (20 mm diameter, 150 µm thickness) with a NiO(Novamet)-YSZ anode and a $(La_{0.8}Sr_{0.2})_{0.95}MnO_3$ (LSM, Praxair)-based cathode were used in this study. The YSZ electrolyte was produced *via* aqueous tape casting and fired at 1450 °C for 3 h. The NiO-YSZ (59.5:40.5 wt.%) cermet anode was screen printed on one side of the electrolyte and fired at 1350 °C for 2 h. The cathode, which comprised of one layer of composite ink LSM-YSZ (50:50 wt.%) and one layer of pure LSM, was also incorporated *via* screen printing and fired at

1100 °C for 2 h. Silver mesh and wires were used as current collectors on both electrodes and fired at 750 °C for 30 min prior to electrochemical testing.

Fuel Composition

The waste composition investigated was representative of those generated in military bases, containing 95 wt.% wood and 5 wt.% polystyrene. A total amount of 2 g of solid fuel mixture was used in each experiment. The materials were ground separately using a hammer mill and then mixed afterwards.

Electrochemical Set-Up and Fuel Cell Testing Protocol

The possibility of using this type of waste as fuel for SOFCs without any pre-treatment was initially evaluated in batch mode using a fixed amount (2 g) of the wood-polystyrene mixture in direct contact with the anode. The setup used for the electrochemical testing is shown in Figure 1. The cell was sealed on an alumina tube using a ceramic sealant (Toku$^{TM}$) with the anode side up. The solid waste mixture was then deposited on top of the anode side. A gas flow rate of 10 ml·min$^{-1}$ of nitrogen was used in the anode side, whereas static air was used in the cathode side. The cell with the solid fuel inside was then heated at a ramp rate of 3 °C·min$^{-1}$ to the first testing temperature, 650 °C. The electrochemical performance was evaluated up to 800 °C, in incremental steps of 50 °C. At each testing temperature, open circuit voltage (OCV) measurements, potential stair-step (from OCV to 0.3 V, 25 mV step size for 5 s) and impedance measurements (50 mV AC amplitude, 100000 to 0.1 Hz) were recorded using a Solartron 1280Z electrochemical interface.

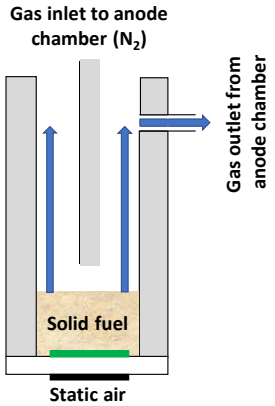


Figure 1. Schematic of the lab-scale rig used for the electrochemical batch testing.

Materials Characterisation

The elemental composition (C, H, N wt.%) of the wood pellets and polystyrene used to create the solid fuel mixture was determined by microanalysis at London Metropolitan University using a Thermo Scientific Flash 2000 Organic Elemental Analyser. Sulphur content was negligible and, therefore, oxygen content was calculated by difference. The

microstructure of the fuel cells, before and after electrochemical testing, was analysed using a Scios Dualbeam focussed ion beam-scanning electron microscope (FIB-SEM). Room temperature powder x-ray diffraction (PXRD) was undertaken with a PANalytical Empyrean diffractometer with Cu-Kα radiation in reflection mode for 2 hours from 10° to 90° 2θ. The obtained PXRD patterns were analysed for phases by using HighScore Plus. The remaining solid after electrochemical testing was analysed *via* PXRD and elemental analysis.

## Results

Figure 2 shows the microstructure of the NiO-YSZ anode and LSM-based cathode before electrochemical testing. Both electrodes had a total thickness of 40 µm. In the case of the LSM-based cathode, each layer (LSM-YSZ and LSM) had a thickness of 20 µm. As seen in Figure 2, some porosity can be observed in the anode electrode after firing. However, most of the porosity would be developed during the *in-situ* reduction.

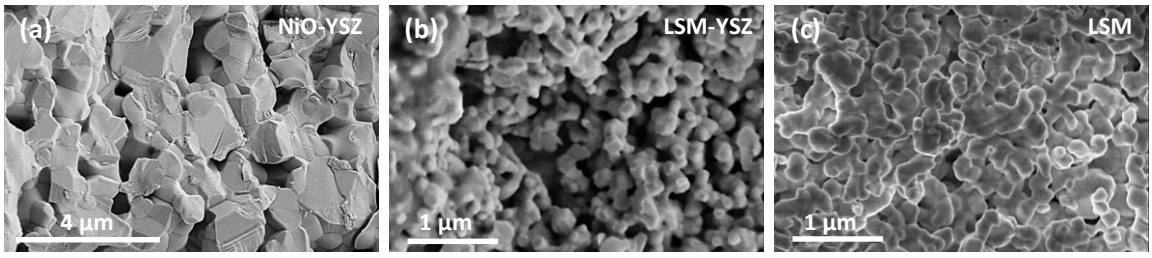


Figure 2. Cross-section micrographs of the as-prepared (a) NiO-YSZ anode and (b, c) LSM-based cathode.

The wood pellets and polystyrene were not subjected to any pre-treatment before being used as fuel, apart from shredding. The sieving of the different fractions of each component showed that the particle size of the polystyrene was between 0.5 and 1 mm. For the wood pellets, 45 wt.% had particle sizes larger than 1 mm, 45 wt.% were between 0.25 and 1 mm and 10 wt.% were lower than 0.25 mm. The elemental analysis showed that the polystyrene was mainly composed of C (~92 wt.%) and H (~8 wt.%), whereas the wood pellets contained a considerable amount of O (~45 wt.%) in addition to C (~49 wt.%) and H (~6 wt.%).

During heating to the working temperature in inert atmosphere, the solid waste mixture decomposes into different fractions, *i.e.* gas ($H_2$, CO, $CO_2$, $CH_4$, *etc.*), oil and solid residue. Both the gas and the solid carbon fractions can act as reducing agents for the anode active phase, in this case NiO. Experiments to measure the $pO_2$ of the solid waste mixture gases were carried out in a separate rig replicating the heating procedure and experimental conditions in which the batch electrochemical testing was carried out (*i.e.* same amount of solid waste, heating ramp rate, dwelling time, *etc.*). Initially, a $N_2$ flow rate of 80 $ml \cdot min^{-1}$ was set for these experiments to guarantee that enough gases were reaching the $pO_2$ sensor and a reading was achievable. The tests were repeated with only 10 $ml \cdot min^{-1}$ of $N_2$ carrier gas, as in the electrochemical batch tests, showing reproducible measurements. For comparison purposes, the $pO_2$ of a dry and humidified $5\%H_2/N_2$ atmosphere was also measured in the same rig following the same experimental protocol as the one used for the solid waste. Table I shows the $pO_2$ values found at the different testing temperatures for the off-gas from the decomposition of pure wood pellets

and a 95 wt.% wood pellets and 5 wt.% polystyrene mixture, together with the two reference gas atmospheres used (dry and humidified 5%$H_2/N_2$). The results show that the atmosphere produced by the solid waste is comparable to a humidified 5%$H_2/N_2$ gas mixture. Although less reducing than dry 5%$H_2/N_2$, it is sufficient to reduce the anode *in-situ* without the incorporation of any other gas which could alter the chemical composition or the decomposition process of the solid waste evaluated.

**TABLE I.** $pO_2$ values (atm) of the different samples tested in function of the temperature.

| | **$pO_2$-658 °C (atm)** | **$pO_2$-700 °C (atm)** | **$pO_2$-750 °C (atm)** | **$pO_2$-800 °C (atm)** |
|---|---|---|---|---|
| **Dry 5%$H_2/N_2$** | $10^{-24.5}$ | $10^{-23}$ | $10^{-21.7}$ | $10^{-20.6}$ |
| **Humidified 5%$H_2/N_2$** | $10^{-22.5}$ | $10^{-21.2}$ | $10^{-20}$ | $10^{-18.8}$ |
| **Wood pellets** | $10^{-22.4}$ | $10^{-21.3}$ | $10^{-20.1}$ | $10^{-19.2}$ |
| **95 wt.% wood + 5 wt.% polystyrene** | $10^{-22.5}$ | $10^{-21.3}$ | $10^{-19.7}$ | $10^{-19.2}$ |

Figure 3 shows the IV and IP curves obtained with 2 g of wood and polystyrene solid fuel mixture (95:5 wt.%) at the different testing temperatures. The data has been corrected by the total active area, 1.16 $cm^2$. The OCV measured were approximately 0.99, 0.98, 0.96 and 0.95 V at 650, 700, 750 and 800 °C, respectively. As can be seen in Figure 3, the maximum power density increased with increasing testing temperature, reaching a value of approximately 62 $mW \cdot cm^{-2}$ at 800 °C, which is encouraging taking into account that no secondary electrolytes, catalysts nor active carrier gases were used during the electrochemical testing. This performance is comparable with the one obtained by Cai *et al.* using gases from pyrolysis-gasification of PP with a Ag-GDC/YSZ/Ag-GDC button cell (71 $mW \cdot cm^{-2}$ at 800 °C) (18). However, in this study the anode was only in direct contact with gases, and not with any oil or solid fractions which can be detrimental to the electrochemical performance of the fuel cell (18).

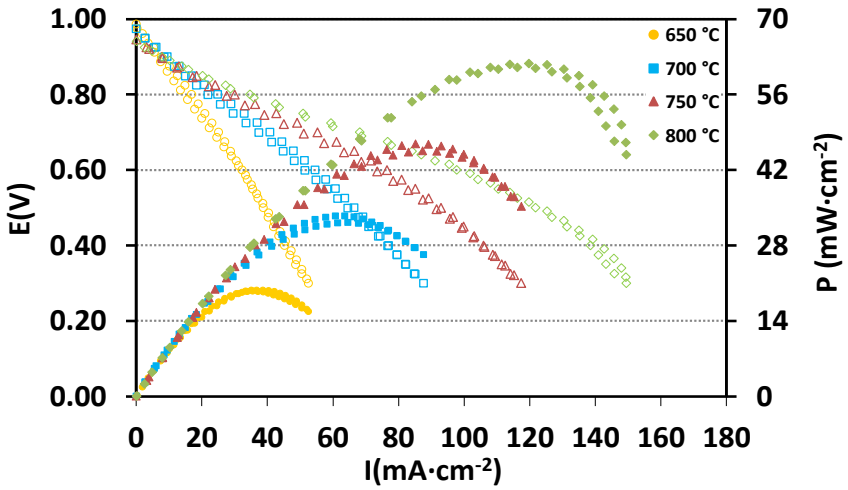


Figure 3. IV and IP curves obtained with the wood+polystyrene mixture at the different testing temperatures using a NiO-YSZ/YSZ/LSM-YSZ/LSM fuel cell.

The complex plane and Bode plot of the AC impedance data collected at the different testing temperatures are shown in Figure 4. As for the IV and IP curves, the data has been corrected by the total active area, 1.16 $cm^2$. The series resistance, $R_s$, decreases with

increasing testing temperature from approximately 1.81 $\Omega \cdot cm^{-2}$ at 650 °C to 0.91 $\Omega \cdot cm^{-2}$ at 800 °C. The total resistance, $R_p$, also decreases with increasing testing temperature, from approximately 11.04 $\Omega \cdot cm^{-2}$ at 650 °C to 3.86 $\Omega \cdot cm^{-2}$ at 800 °C. Studies with symmetrical cathode cells were performed to evaluate the cathode contribution for this cell configuration. It was found that the resistance for the LSM-YSZ/LSM cathode decreased from 1.90 $\Omega \cdot cm^{-2}$ at 650 °C to 0.13 $\Omega \cdot cm^{-2}$ at 800 °C and, therefore, contributing to 17% and 3.4% of the $R_p$, respectively. In Figure 4 (b) can be seen that at the lower testing temperature, 650 °C, there is a large contribution at around 100 Hz. With increasing testing temperature, up to 800 °C, a clear secondary arc becomes resolved at a higher frequency domain, between 2000 to 4000 Hz.

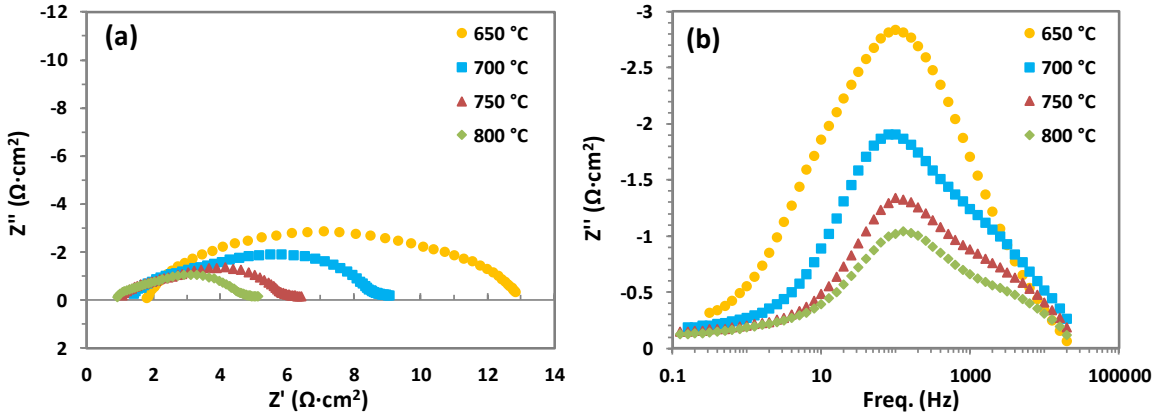


Figure 4. The (a) complex plane and (b) Bode plot of the AC impedance data recorded at OCV for the NiO-YSZ/YSZ/LSM-YSZ/LSM fuel cell operated with a wood+polystyrene mixture at the different testing temperatures.

Figure 5 shows a comparison of the PXRD patterns of the as-prepared *vs.* the *post-mortem* NiO-YSZ electrode. The results show that *in-situ* reduction of the NiO to Ni took place during the electrochemical testing. The *post-mortem* XRD presents some contributions from Ag and $Al_2O_3$ related to the current collectors and sealant, respectively, as well as some unknown contributions between 20° and 30° (marked with * in Figure 5). These unknown contributions were only observed in some of the electrochemical tests, independently of the anode material electrode and, therefore, they are more likely related to impurities from the sealing.

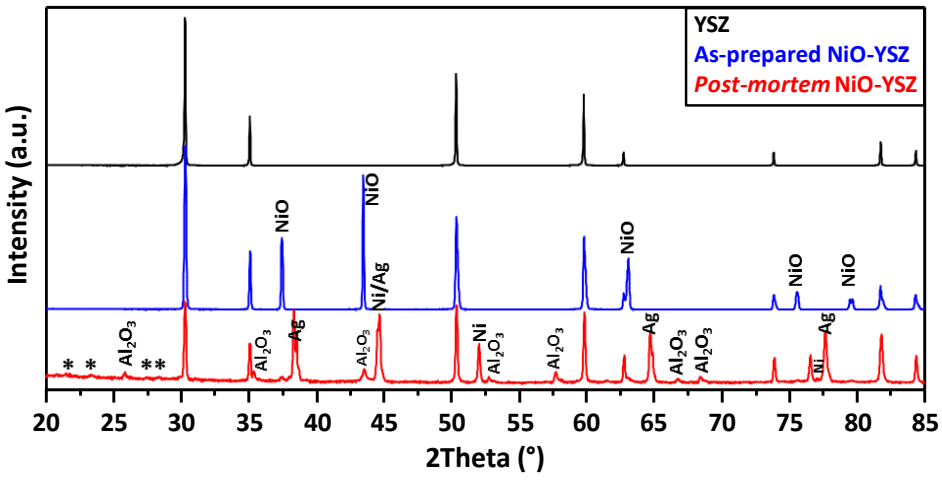

Figure 5. Comparison of the PXRD patterns of the NiO-YSZ anode before and after electrochemical testing with a wood+polystyrene mixture from 650 to 800 °C.

The SEM studies of the *post-mortem* anode showed that upon *in-situ* reduction of NiO to Ni during the electrochemical testing (as proven by the PXRD pattern shown in Figure 5), the anode porosity was further developed. Figure 6 shows a cross-section SEM micrograph of the NiO-YSZ anode after testing. Some long filaments that are bridging together, which could be carbon deposits, were identified. As explained previously, during heating the fuel mixture decomposes into gas, oils and a remaining solid. The GC analysis of the outlet gas collected during the electrochemical testing showed the presence of $H_2$, CO, $CO_2$, $CH_4$ and some $C_2$ hydrocarbons up to 700 °C. At 750 and 800 °C the $CH_4$ and $C_2$ levels decreased significantly. The diffusion of hydrocarbon gas species, and probably some of the oil fraction, through the porous anode electrode is plausible and, therefore, it would not be surprising to find some carbon deposits on the Ni-based anode in these conditions.

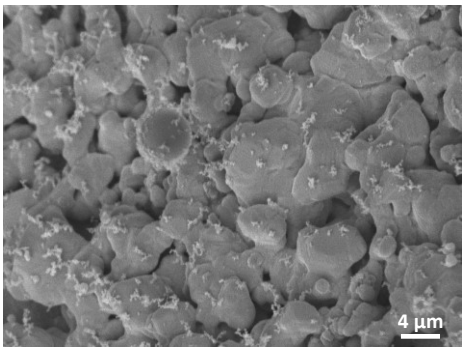


Figure 6. Cross-section SEM micrograph of the NiO-YSZ anode after electrochemical testing with a wood+polystyrene mixture from 650 to 800 °C.

The PXRD analysis of the remaining solid waste after electrochemical testing showed the three characteristic reflections of amorphous carbon at 25°, 44° and 83° (Figure 7). Elemental analysis confirmed that the remaining solid is mostly composed of C (~82 wt.%) with small proportions of H (~1wt.%) and O (~14 wt.%). Compared to the initial solid waste mixture, the remaining solid is enriched in C, most likely due to the release of most of the volatile functional groups at the high testing temperatures.

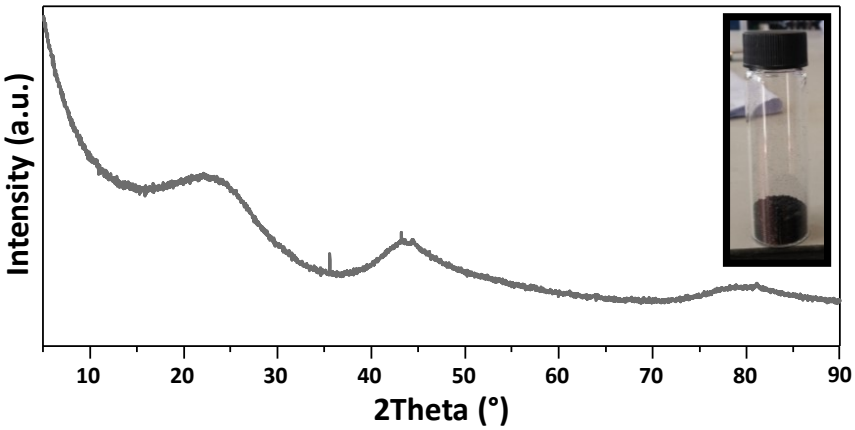

Figure 7. PXRD pattern of the remaining solid fraction from the wood+polystyrene mixture after electrochemical testing from 650 to 800 °C (inset. an image of the residual solid waste collected after the electrochemical test).

## Conclusions

The electrochemical evaluation of a wood and polystyrene mixture as fuel was carried out in batch testing mode (a fixed amount of solid waste was in direct contact with the anode) using a 150 μm YSZ supported cell with a NiO-YSZ reference anode and LSM cathode. These initial results show the feasibility of using this complex solid waste as fuel in SOFCs without any pre-treatment, and without the aid of secondary electrolytes or catalysts mixed into the fuel. In these conditions, a maximum power density of 62 $mW \cdot cm^{-2}$ was achieved at 800 °C. The improvement of the electrochemical performance for long-term operation can be achieved with the use of alternative anode materials that are coking resistant and highly catalytic active, not only towards $H_2$ but also CO and hydrocarbons. Results with alternative ceramic materials for this type of solid waste fuels, and the development of an integrated SOFC system that works with a continuous feeding of solid waste are under investigation.

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